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# NONLINEAR OPTICAL THIN FILMS


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## Final Report



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The effort consisted of experimental work to deposit and characterize thin films as well as a modeling effort. PZLT materials were emphasized, with several compositions examined. In addition, limited effort was directed toward deposition of other materials, including barium titinate and strontium-barium niobate.

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## 1.0 INTRODUCTION

This is the Final Report for the effort "Nonlinear Optical Thin Films" for the period 1 October, 1988 through 29 December 1990. The effort consisted of experimental work to deposit and characterize thin films as well as a modeling effort. PLZT materials were emphasized, with several compositions examined. In addition, limited effort was directed toward deposition of other materials, including $BaTiO_3$ and $(Sr,Ba)Nb_2O_6$.

## 1.1 EXPERIMENTAL INVESTIGATION

The experimental effort included RF magnetron deposition of PLZT of 7/0/100, 8.6/0/100, 28/0/100, 7.6/70/30 compositions. Additionally, ion beam sputtering was used to deposit 28/0/100, 15/0/100, 7/0/100, and 0/0/100. Many substrate materials were used, including fused silica, sapphire, Si, and Si and GaAs with buffer layers of $SiO_2$. A major thrust of the ion-beam deposition effort was the growth of films with good crystallinity and orientation on GaAs substrates at temperatures below $\sim$500 °C This is necessary to prevent degradation of the GaAs material and of any device structures which may be present. This will be important in moving towards integrated PLZT/GaAs photonic devices. This goal was achieved.



We used the following diagnostic techniques to evaluate the films:

X-ray diffraction to determine the crystal structure

Electron dispersion spectroscopy to determine the elemental composition

Alpha-step to determine film thickness

Confocal scanning differential polarization microscopy to determine the film electrooptic properties

Second harmonic generation was obtained using YAG laser output at 1.06 µm.as the fundamental wavelength. We investigated the angular dependence of the intensity of the SHG from the films to determine components of the nonlinear optical tensor.

Waveguiding was achieved in PLZT in structures of ITO/PLZT/$SiO_2$/Si and ITO/PLZT/$SiO_2$/GaAs. By applying an electric field of 0 to 300 V/mm to the ITO and Si substrate, and using prism out-coupling, we observed modal angular shifts. The total angular shift was due primarily to the contribution from a positive change in the effective index of the film which was likely the result of changing the thickness of the film. The contribution from pure electric-field-induced birefringence change is small and reduces the index. Both TE and TM modes exhibit similar shifting behaviour.

We investigated the time evolution of the carrier distribution and induced electric field in a photorefractive material in the presence of a cw laser field and a pulsed electric field. Some effort was spent investigating the effects of the photovoltaic term in both the steady state and the time dependent situations. In addition, we investigated the high-speed switching characteristics of PLZT and observed the material to a response time of less than 2 nsec.

Plasma etching was examined using chlorinated and fluorinated plasmas produced using instrumentation-limited dc hollow cathode discharges. Etch rates of PLT thin

films as high as 650 nm/hour were obtained. After etching, the stoichiometry was characterized to indicate that in some situations (e.g. elevated substrate temperature) significant changes occur. This is significant if the material is to be applied to construct electronic and optoelectronic devices.

## 1.2 THEORETICAL INVESTIGATION

The theoretical efforts were a relatively small portion of this project. The modeling was carried out by Chibing Xu, under the supervision of Professor McIver and constituted a major portion of his PhD dissertation. This work has also resulted in two completed manuscripts that have been submitted for publication in the Journal of the Optical Society of America B.

The subject of these manuscripts is the response of a photorefractive material to a cw gaussian laser beam and a transverse electric field. This work was motivated by the frequency doubling experiments performed in the Center for High Technology Materials on thin film PLZT materials. We were particularly interested in the response of the photogenerated carriers to a time-dependent externally applied electric field.

The first paper explores the steady state behavior of a photorefractive material subjected to a cw gaussian laser beam. The photorefractive material is described by the phenomenological band transport model of Kukhtarev. In order to simplify the mathematics the calculations were carried out in one dimension and the photovoltaic term was ignored. The latter approximation is both common and justified for most materials. Unlike other calculations, we did not linearize the equations. As can be seen in the attached manuscript the nonlinear terms can be important at relatively low intensities for some materials.

The second paper extends the results of the first paper by adding a transverse electric field to the material. In some cases this field is periodically turned on and off thereby simulating the conditions under which the frequency doubling experiments were performed. These calculations showed that the external electric field in the illuminated region of the material was reduced because of the external field, that the relaxation time of the elctric field within in the materials depends strongly on the intensity of the light, and that the presence of an oscillating electric field can enhance the induced electric field.

## 2.0 PUBLICATIONS

The following publications resulted from this effort:

1. C. Xu, D. Statman, and J. K. McIcer, "Effects of a Gaussian Beam on Photorefractive Materials: I. Steady State Solution", Submitted to Journal of Optical Society of America B.

2. C. Xu, J. K. McIver, and D. Statman, "Effects of a Gaussian Beam on Photorefractive Materials: II. Dynamic Behavior Under Externally Applied Electric Field", Submitted to Journal of Optical Society of America B.

3. F. Wang, C.-B. Juang, C. Bustamante, and A. Y. Wu, "Electro-Optic Properties of $(Pb,La)(Zr,Ti)O_3$, $BaTiO_3$, $(Sr,Ba)Nb_2O_6$ and $Ba_2NaNb_5O_{15}$ Thin Films by a Confocal

Scanning Differential Polarization Microscope", in the 4th International SAMPE Electronics Conference, Volume 4, Electronic Materials-Our Future, pp. 712-721, ed. R. E. Allred, R. J. Martinez, and K. B. Wischmann, (Society for the Advancement of Materials and Process Engineering) 1990.

4. A. Y. Wu, "Deposition of (Pb,La)(Zr,Ti)$O_3$, $BaTiO_3$, (Sr,Ba)$Nb_2O_6$, $Ba_2NaNb_5O_{15}$, $KTiOPO_4$, and beta-$BaB_2O_4$ Thin Films", in the 4th International SAMPE Electronics Conference, Volume 4, Electronic Materials-Our Future, pp. 722-733, ed. R. E. Allred, R. J. Martinez, and K. B. Wischmann, (Society for the advancement of Materials and Processing Engineering) 1990.

5. M. R. Poor, A. M. Hurd, C. B. Fleddermann, and A. Y. Wu, "PLZT Thin Film Etching using Plasma Techniques", in the Seventh International Symposium on the Applications of Ferroelectrics, University of Illinois, Urbana, Illinois (1990). (In press)

6. A. Y. Wu, F. Wang, C.-B. Juang, and C. Bustamante, "2-D High Definition and High Resolution PLZT Thin Film Spatial Light Modulators", in the Seventh International Symposium on the Applications of Ferroelectrics, University of Illinois, Urbana, Illinois (1990). (In press)

7. A. Y. Wu, F. Wang, C.-B. Juang, C. Bustamante, C.-Y. Yeh, and J.-C. Diels, "Electro-Optic and Non-Linear Optical Properties of (Pb,La)(Zr,Ti)$O_3$, $BaTiO_3$, (Sr,Ba)$Nb_2O_6$, $Ba_2NaNb_5O_{15}$, and beta-$BaB_2O_4$", in the Seventh International Symposium on the Applications of Ferroelectrics, University of Illinois, Urbana, Illinois (1990). (In press)

8. F. Wang and A. Y. Wu, "Electro-Optical and Nonlinear Optical Properties of Thin Film Materials Containing Oxygen-Octahedra Under High DC Electric Field", in the Seventh International Symposium on the Applications of Ferroelectrics, University of Illinois, Urbana, Illinois (1990). (In press)

9. A. Mukherjee, S. R. J. Brueck, and A. Y. Wu, "Electric Field Induced Second Harmonic Generation in PLZT", Opt. Commun. 76(3,4) 220 (1990).

10. A. Y. Wu, F. Wang, C.-B. Juang, and C. Bustamante, "Electro-Optic and Non-Linear Optical Coefficients of (Pb,La)(Zr,Ti)O3, BaTiO3, (Sr,Ba)Nb206, and Ba2NaNb$_5$015 Thin Films", in the Symposium on Ferroelctric Thin Films, Spring Meeting, Materials Research Society, San Francisco, California (1990). (In press)

11. M. R. Poor, A. M. Hurd, C. B. Fleddermann, and W. Y. Wu, "Plasma Etching of PLZT and PLT using Chlorine Based Gases", in the Symposium on Ferroelectric Thin Films, Spring Meeting, Materials Research Society, San Francisco, California (1990). (In press)

12. L.L. Boyer, A. Y. Wu, and J. R. McNeil, "Physical and Electro-Optical Properties of Ion Beam Sputtered Thin Film PLZT Ceramic", in the Symposium on Ferroelectric Thin Film, Spring Meeting, Materials Research Society, San Francisco, California (1990). (In press)

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16. Feiling Wang and A. Y. Wu, "An Analytical Model for the Quadratic Electrooptic Effect of Perovskites", Submitted to Physical Review Letters.

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